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Alternative layouts for the carbon capture with the Chilled Ammonia Process

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Abstract

Many alternatives are being investigated for the carbon capture, but none appears to have been proved as the choice for full-scale applications. This work considers the Chilled Ammonia Process for coal-fired Ultra Super Critical power plants. Three layouts are simulated with Aspen Plus and the Extended UNIQUAC thermodynamic model. Compared to a traditional layout, stripping of the wash water of the absorber or, better, splitting the rich solution between the middle and the top of the column limits greatly the ammonia slip. Moreover, splitting the regeneration over two levels reduces substantially the electric loss due to stream extraction from the turbine. The simulations show that the net electric efficiency drops from 45.5% to 33.5-34.5%, the SPECCA index is 3.8-4.3 $MJ_{th}\ kg_{CO2}^{-1}$ and the heat duties are 2.7-2.9 $MJ_{th}\ kg_{CO2}^{-1}$. The performances may improve greatly upon optimization of the parameters.

Keywords: carbon capture; chemical absorption; ammonia aqueous solution; chilled ammonia process; process layouts.

1. Introduction

Many alternatives are being investigated worldwide to capture, and then store, the carbon dioxide generated by the combustion of fossil fuels. Apparently none has been proved to be the choice for full-scale applications. This work considers the post-combustion chemical absorption via an aqueous solution of ammonia in chilled conditions, the Chilled Ammonia Process (CAP), applied to coal-fired Ultra Super Critical (USC) power plants. The scope is comparing three layouts with the software Aspen Plus (ver.7.3) employing a thermodynamic model that is not built inside the code but defined by the user.

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Nomenclature

Equipment abbreviations		ST	Stripper
AB	Absorber	WK	Water knockout
AC	Air-cooler	WT	Wash tower
CC	Contact cooling tower	*Acronyms*	
CM	Compressor	CAP	Chilled Ammonia Process
FGD	Flue gas desulfurization	FGD	Flue Gas Desulfurization
FN	Fan	USC	Ultra Super Critical
HC	Hydrocyclone	*Symbols*	
HX	Heat exchanger	η_{CO2}	Carbon capture efficiency [-]
PM	Pump	η_e	Net electrical efficiency [-]
PR	Purge	E	Specific CO_2 emission [kg_{CO2} MWh_e^{-1}]
RB	Reboiler	$SPECCA$	Specific Primary Energy Consumption
RC	Recuperator		for Carbon Avoided [MJ_{th} $kgCO_2^{-1}$]
RG	Regenerator	q_{CO2}	Specific heat duty [MJ_{th} kg_{CO2}^{-1}]

The adopted thermodynamic model, namely the Extended UNIQUAC model, has been developed over the years at the Technical University of Denmark for diverse mixtures, including amines and ammonia solutions for the carbon capture. It has been tuned finely for reproducing phase equilibria and thermal properties of the CO_2-NH_3-H_2O system [1-4]. On its side, Politecnico di Milano has been focusing mainly on plant schemes adopting a simplified thermodynamic model [5] or the Aspen Plus V7.2 built-in e-NRTL model [6,7]. The two universities have naturally joined in the study of post-combustion carbon capture via chemical absorption. At first, Darde et al. [8] have shown that the Extended UNIQUAC model is averagely more accurate than the e-NRTL model in reproducing equilibrium experimental data of the ternary system CO_2-H_2O-NH_3 over a wide range of temperatures, pressures and concentrations. The joint analysis covers here the comparison of capture processes that, as in the past, involve salt precipitation and that, consequently, are simulated with an equilibrium-stage approach.

2. Bibliographic review

The first conceptual scheme of a carbon dioxide chemical absorption with aqueous ammonia is likely that by Bai et Yeh (1997) [9]. It is a conventional scheme which is envisioned to have a water wash at the top of both the absorber and the regenerator because ammonia slip is already recognized as a possible problem. Resnik et al. (2004) [10] are probably the first investigators to suggest the use of ammonia solution for the multi-pollutant (CO_2, SO_2, NO_x, HCl and HF) from the flue gases of fossil fuel-fired plants, an idea that is being pursued by Powerspan Corp [11]. In 2005 Gal patents the concept of conducting the absorption in chilled conditions (0-20°C) to favor the carbon dioxide capture and to limit the ammonia slip [12]. The company Alstom has licensed the exclusive, world-wide rights to market and sell the process patented by Gal [13]. Until about 2009 Alstom designs and operates a pilot plant based on the conventional absorption-regeneration scheme in which, though, the regeneration pressure is fairly high (20-40 bar). Subsequently, Alstom redesigns the layout modifying the way ammonia is recovered from the flue gas and implements it in a few test sites [14-16]. Strube and Manfrida (2011) [17] study a capture layout similar to this second Alstom scheme and compare its integration with different power plant types. Also starting from the later scheme by Alstom, Linnenberg et al. (2012) [18] develop two alternative arrangements of the absorption stage and analyze in detail the integration with the power plant. Finally, there is quite a number of patents deposited by Alstom that cover many modifications to its layouts, but to the knowledge of the authors there are no scientific publications about them yet.

3. Thermodynamic model and process layouts

The USC power plant equipped with the CAP is divided into two major blocks: (i) the power and (ii) the capture block. The power block is treated as a whole, whereas the capture block is subdivided into islands: (i) exhaust chilling, (ii) absorption-regeneration-gas wash, (iii) carbon dioxide compression, (iv) chilling plant, and (v) ammonia removal. Three layouts for the capture block are considered, differing primarily by the abs.-reg.-gas wash island. The ammonia removal island is not present in one layout because the ammonia slip is controlled at the absorption process. General parameters are given in Table 1.

Extended UNIQUAC model

The Extended UNIQUAC thermodynamic model for gas solubility in salt solutions was developed by Thomsen and Rasmussen (1999) [1]. It is derived from the original UNIQUAC expression by Abrams and Prausnitz (1975) [20] by adding a Debye-Hückel term to account additional excess Gibbs energy from the electrostatic interactions between ionic species. The model requires UNIQUAC volume and surface area parameters for each species, along with temperature-dependent binary interaction energy parameters for each pair of species. Phase equilibrium calculations are performed with the $\gamma - \varphi$ approach coupled with equilibrium speciation reactions with potential solid phase precipitation. The liquid phase activity coefficients are calculated from the Extended UNIQUAC model, while the gas phase fugacity coefficients from the Soave-Redlich-Kwong equation of state. Besides phase relations, the model reproduces also thermal properties, such as enthalpy and entropy, within the experimental accuracy.

Power block

The effect of the steam extraction on the power generation is computed starting from a typical expansion curve of a low pressure turbine. The curve is assumed to be a straight segment connecting inlet and outlet of the turbine on an entropy-enthalpy diagram (Figure 2). The extraction pressure along the curve is determined by the regeneration temperature allowing for a minimal temperature difference in the reboiler. Prior to entering the reboiler, the steam is attempered with part of the liquid water exiting the reboiler itself. The extracted mass flow rate is defined by the energy balance over the reboiler for a given heat duty. The electric loss due to the steam extraction is computed as the power that would be generated by the extracted steam from the extraction state to the outlet state assuming that the expansion curve does not change. The integration of the exiting condensate with the power block, such as in the deaerator or in the pre-heating line, is not considered now. The condensate is instead directed to the condenser.

Capture block

Layout 1 is derived from the first plant proposed by Altsom [13] and based on a conventional absorption-regeneration scheme (Figure 1). Such scheme applied to CAP has the major issue of the ammonia slip through the treated flue gas requiring: (i) refrigeration, (ii) water wash and (iii) removal of remaining ammonia with an acid solution. The ammonia slip issue may be addressed at the design of both the absorber and the process. Regarding the absorber, Budzianowski [19] investigates numerically and, in part, experimentally three reactor configurations and shows that a number of parameter may be adjusted to control the vaporization of ammonia. Regarding the process, new schemes may be defined.

Layout 2 is the evolution of Layout 1 derived from the second plant by Alstom [14] (Figure 1). It aims at recovering the ammonia slip from the treated gas through a high-pressure and high-temperature thermal stripping of a small portion of the wash water of the absorber.

Layout 3 is a modification of Layout 1 that aims at controlling the ammonia slip, as Layout 2 but with another strategy, and the electric loss due to stream extraction (Figure 1). The ammonia slip is limited splitting the recycle of the rich solution between the top and the middle of the absorber; the electric loss is limited splitting the regeneration into two stages, one at low pressure and low temperature and the other at high pressure and moderate temperature. Pressures and temperatures are set imposing that the two compressors have same compression ratio and same inlet volume flow rates so that they are identical.

Performance indexes

The carbon capture efficiency, η_{CO2} [%] defined as the ratio of the flow rates [kmol s^{-1} or kg s^{-1}] of the carbon dioxide exiting the compression island and that entering the exhaust chilling island, is one performance index. The second one is the specific heat duty, q_{CO2} [MJ_{th} kg_{CO2}^{-1}], defined as the ratio of the reboiler heat duty [MW_{th}] and the mass flow rate [kg s^{-1}] of the captured carbon dioxide. However, the specific heat duty does not include the information on the capture efficiency nor on the temperature at which the heat duty is required (or in equivalent terms the loss of electric power from the steam turbine). Thus, here is adopted a third index, the Specific Primary Energy Consumption for Carbon Avoided ($SPECCA$) [MJ_{th} kg_{CO2}^{-1}], introduced by Campanari et al. [21] and in use by the authors [7,8]. As an indication, the $SPECCA$ of a conventional MEA plant exceeds 4 MJ kg_{CO2}^{-1} [22].

Design parameters

The design parameters are: (i) ammonia initial concentration of the aqueous solution, (ii) ammonia-to-carbon dioxide ratio in the absorber, (iii) regeneration pressure, and (iv) regeneration temperature. The ammonia-to-carbon ratio in the absorber is the ratio of the ammonia moles entering the reactor through the lean solution and the carbon dioxide moles entering through the exhaust. For each layout the design parameters are varied to: (i) achieve the carbon capture efficiency η_{CO2} of 90%, (ii) reduce the ammonia slip in the treated gas and in the compressed carbon dioxide respectively below 100 mg m^{-3} at normal condition and at 6% of O_2 and below 5 ppm, both on a dry basis, and (iii) limit the lean solution flow rate. The chosen values are in Table 2. With respect to a previous work [7], which minimizes the $SPECCA$, the attention is here on the ammonia slip. In the future, an optimization will be conducted.

4. Results and discussion

The ratio of the electric power loss due to the stream extraction from the turbine and the heat duty as a function of the regeneration temperature is depicted in Figure 2. At a temperature as low as 80°C a heat duty of 1 MW_{th} corresponds to a power loss of 0.119 MW_e. This loss grows rapidly to 0.166 at 100°C, 0.212 at 120°C and 0.256 MW_e at 140°C. At the stripping temperature of 200°C it is already 0.377 MW_e.

Table 3 reports the results in terms of the electric power loss due to direct use by air-coolers, fans, compressors and pumps, or indirect use by heat exchangers (via the chilling plant) and reboilers (via the extracted steam). The exhausts chilling island is identical for all layouts and consumes the least amount. Air-coolers and pumps of the absorption-regeneration-gas wash island sum to a relatively small portion of the consumptions, especially for Layout 2 that has small flow rates due to higher ammonia concentrations. Heat duties and chilling loads account for most of the losses. Layout 2 shows the lowest loss due to chilling thanks to a reduce load on the recycle of the rich solution. In addition, Layout 2 and, in particular, Layout 3 prove a greatly reduced loss due to the steam extraction. On top of this, Layout 3 allows to control effectively the ammonia slip as demonstrated by the smallest consumption for the pump of the water wash of the absorber (PM23 in Figure 1) and the absence of the ammonia removal island.

Losses are between 180 and 200 MW_e for a USC plant of 754 MW_e (Table 4). The net electrical efficiency goes from 45.5% to about 33.5-34.5%. $SPECCA$ values are higher than a previous work [7]: Darde et al. [8] show that performances simulated with the Extended UNIQUAC model, as here, are worse than with the e-NRTL model, as before. Furthermore, the design parameters must be optimized.

Table 1. General parameters.

Parameter	Unit	Value	Parameter	Unit	Value
Air coolers			*Heat exchangers*		
Fluid end temperature	°C	25	Minimum temperature difference	°C	5
Relative pressure drop	%	1	*Low pressure steam turbine*		
Specific electric consumption	$MW_e\,MW_{th}^{-1}$	0.0159	Inlet pressure	bar	4.5
Ambient air			Inlet temperature	°C	306
Temperature	°C	15	Outlet pressure	bar	0.05
Chilling plant			Outlet vapor title	%	93
Coefficient of performance	$MW_{th}\,MW_e^{-1}$	5	Outlet velocity	m s^{-1}	250
Specific electric consumption	$MW_e\,MW_{th}^{-1}$	0.20	Generator efficiency	%	98
Columns			*Motors*		
Contact cooler pressure drop	bar	0.01	Electro-mechanical efficiency	%	95
Other column pressure drop	bar	0.03	*Pumps*		
Compressors			Hydraulic efficiency	%	85
Isentropic efficiency	%	85	*Reboiler*		
Last compressor end pressure	bar	80	Steam superheated temperature	°C	5
Fans			Steam subcooled temperature	°C	0
Forced fan end pressure	bar	1.08	*Reference power plant* [22 Sec. 3.2]		
Induced fan end pressure	bar	1.04	Net electric power	MW_e	754
Isentropic efficiency	%	90	Net electrical efficiency, $\eta_{e,REF}$	%	45.5
Exhausts [22 Sec. 3.2]			Specific CO_2 emission, E_{REF}	$kg_{CO2}\,MWh_e^{-1}$	763
Mass flow rate	kg s^{-1}	782	*Pipeline*		
Pressure	bar	1.04	Delivery pressure	bar	110
Temperature	°C	50	*Targets*		
Composition:	% (vol. wet)		Carbon capture efficiency, η_{CO2}	%	90
CO_2		13.73	Max ammonia slip (vol. dry)		
Inert (Ar, N_2,O_2)		76.54	Treated gas	mg $Nm^{-3}{}_{6\%O2}$	100
H_2O		9.73	Compressed carbon dioxide	mg Nm^{-3}	10

Table 2. Adopted design parameters.

Parameter	Unit	Layout 1	Layout 2	Layout 3
Ammonia initial concentration	%wt	0.1	0.2	0.1
Ammonia-to-carbon dioxide ratio	kmol $kmol^{-1}$	3.1	3.2	3.1
Regeneration pressure	bar	20	20	10.7/29.3
Regeneration temperature	°C	110	102/204	69/120

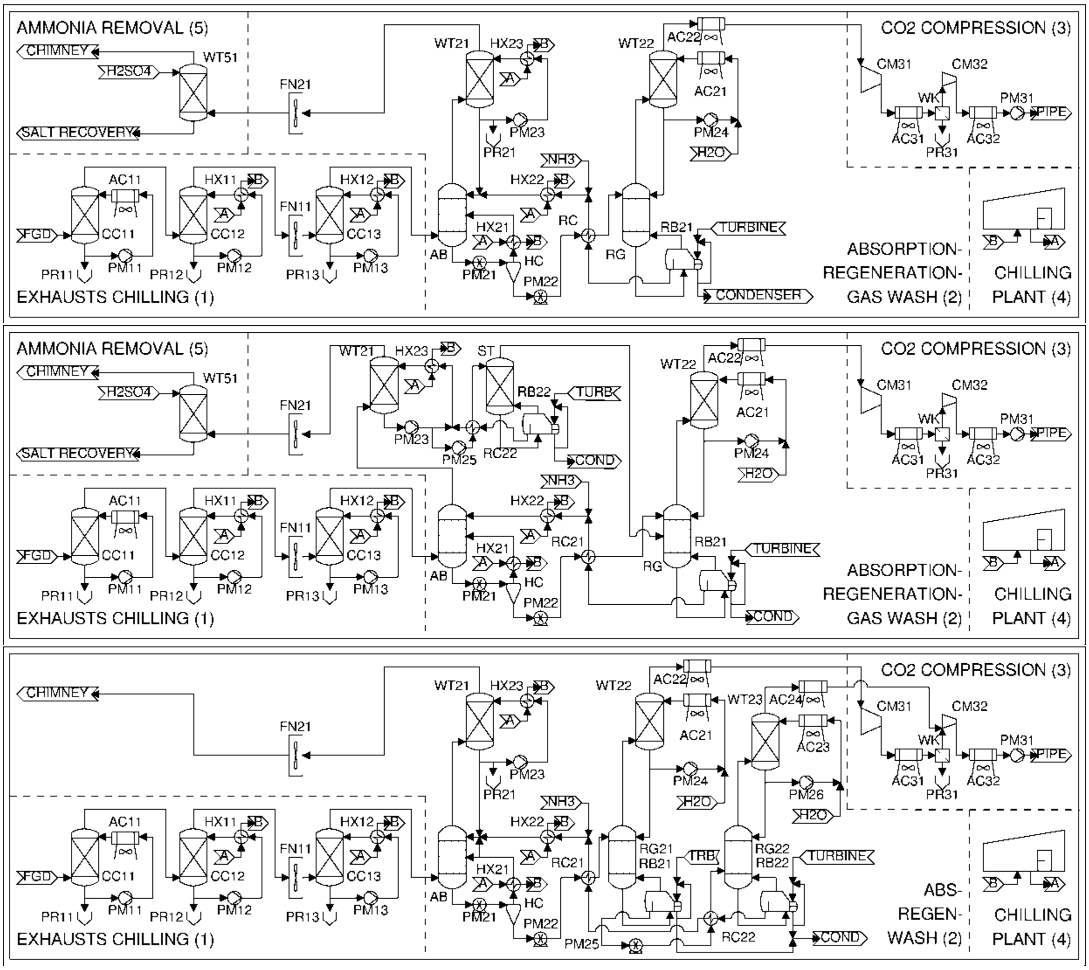

Figure 1. The three layouts considered in this work. From top to bottom: Layout 1, Layout 2 and Layout 3.

5. Conclusions

Three layouts for the carbon capture with the Chilled Ammonia Process (CAP) are compared yielding the following conclusions.

- The ammonia slip is a major issue, but it can be controlled at the process level by either stripping the water wash or, better, by splitting the rich solution between the top and the middle of the absorber.
- Heat duty is the greatest source of electrical loss by way of steam extracted by the turbine, but it may be effectively mitigated by splitting the regeneration process into a low and a high pressure stage.
- CAP is predicted to reduce the net electric efficiency of an USC plant from 45.5% to about 33.5-34.5%, while the SPECCA is 3.8-4.3 $MJ_{th}\ kg_{CO2}^{-1}$ and heat duties 2.7-2.9 $MJ_{th}\ kg_{CO2}^{-1}$.
- Indexes computed in this work are less promising than in previous studies because the Extended UNIQUAC model is expected to be more accurate but less optimistic than the e-NRTL model, built-in inside Aspen Plus, and because the design parameters must be optimized.
- The design parameter optimization must be constrained to limit the ammonia slip.

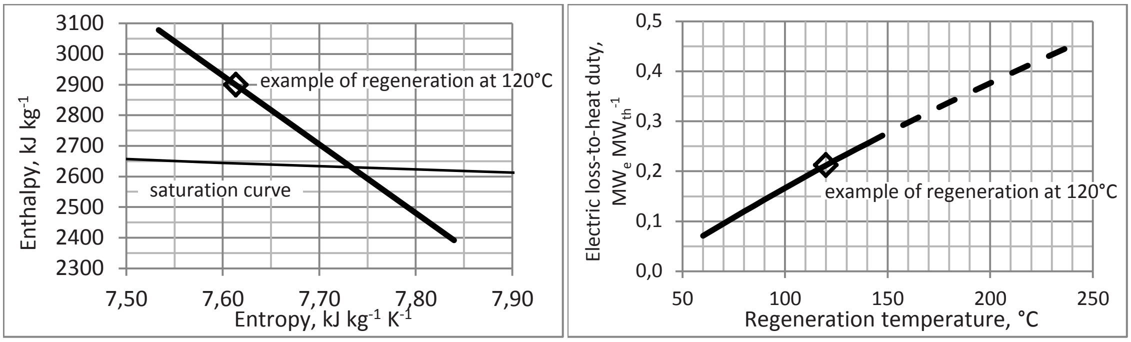

Figure 2. Left: expansion curve in the enthalpy-entropy diagram of the low pressure turbine from which the steam is extracted. Right: ratio of electrical loss-to-heat duty as a function of regeneration temperature (above 140°C the curve is extrapolated because the computed pressure is higher than the inlet pressure). The diamond shows an example of a regeneration temperature at 120°C.

The next stage will consider capturing the slipped ammonia to produce fertilizers and employing a Ljungström heat exchanger to recuperate heat. Moreover, the complete plants will be also optimized.

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Table 3. Electric power loss due to direct (air-coolers, compressors, fans, pumps) and indirect use (heat exchangers, reboilers).

Electric power, MW_e	Layout 1	Layout 2	Layout 3	Electric power, MW_e	Layout 1	Layout 2	Layout 3
Exhaust chilling				*Carbon dioxide compression*			
AC11	1.63	1.63	1.63	AC31	0.16	0.16	0.11
FN11	3.79	3.79	3.79	AC32	0.53	0.53	0.62
PM11	0.21	0.21	0.21	CM31	6.85	6.82	5.51
PM12	0.14	0.14	0.14	CM32	6.16	6.14	9.91
PM13	0.03	0.03	0.03	PM31	0.77	0.77	0.77
Subtotal	5.80	5.80	5.80	Subtotal	14.48	14.42	16.92
Absorption-Regeneration-Wash				*Chilling plant*			
AC21	0.10	0.11	0.03	HX11	7.88	7.88	7.88
AC22	0.06	0.02	0.00	HX12	0.81	0.81	0.81
AC23	0.00	0.00	0.19	HX21	53.46	49.36	54.39
AC24	0.00	0.00	0.01	HX22	26.08	26.99	29.50
PM21	1.12	0.57	1.12	HX23	3.29	1.94	1.08
PM22	4.72	2.70	2.29	Subtotal	91.52	86.98	93.65
PM23	3.81	0.97	2.26	*Power block*			
PM24	0.40	0.07	0.06	RB21	77.60	62.16	28.61
PM25	0.00	0.20	5.18	RB22	0.00	10.97	26.17
PM26	0.00	0.00	0.08	Subtotal	77.60	73.13	54.78
Subtotal	10.21	4.64	11.23	Total of electric power losses	199.61	184.98	182.38

Table 4. Electric performance of the reference plant [22 Sec. 3.2] and the considered layouts.

Parameter	Unit	Reference	Layout 1	Layout 2	Layout 3
Electric power loss	MW_e	NA	200	185	182
Net electrical power	MW_e	754	555	569	572
Net electrical efficiency, η_e	%	45.5	33.5	34.3	34.5
Capture efficiency, η_{CO2}	%	NA	90.5	90.2	89.9
Specific CO_2 emission, E	$kg_{CO2}\,MWh_e^{-1}$	763	98.6	99.5	101.5
Specific heat duty	$MJ_{th}\,kg_{CO2}^{-1}$	NA	2.80	2.48/0.19	2.09/0.85
$SPECCA$	$MJ_{th}\,kg_{CO2}^{-1}$	NA	4.3	3.9	3.8